

Common liptinitic constituents of Tertiary coals from the Bintulu and Merit-Pila coalfield, Sarawak and their relation to oil generation from coal

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Abstract: The onshore and offshore Tertiary sedimentary sequences of Sarawak contain numerous coal seams of Oligocene and Lower Miocene age. The offshore extensions and equivalents of these onshore basins contain oil that is considered to be possibly sourced from these coals and related terrestrial-derived organic matter. Despite this, very little information has been published on the geochemical and petrographic characteristics of the Sarawak coals. To rectify this, all the major Tertiary coal deposits of Sarawak have been visited and samples collected. In this preliminary study, results from two areas i.e. the Bintulu area and the Stapang block of the Merit-Pila coalfield will be presented emphasizing on the type of common liptinitic constituents of the coals and their relation to oil generation and expulsion. A number of petrographic features that are commonly considered to indicate oil generation and expulsion from coals were observed. Such features include the occurrence of exsudatinitite; oil droplets and oil haze; changes of fluorescence intensity; and development of micrinite. The precursors for the exsudatinitite in the Bintulu coals is mainly bituminite and that from Stapang, Merit-Pila is mainly suberinite. The development of the exsudatinitite is taken to represent an early phase of liquid hydrocarbon generation and its expulsion took place at about 0.4% R_o for the Merit-Pila coals and 0.45% R_o for the Bintulu coals (earlier than the generally considered "oil-window" of 0.5% vitrinite reflectance). It has been possible to recognize the order of liquid hydrocarbon generation for a number of the common liptinitic constituents present in these Tertiary coals of Sarawak.

INTRODUCTION

The mid 80's was the time when an increasing number of workers in the field of petroleum geochemistry (e.g. Durand and Paratte, 1983; Bertrand, 1984; Thompson *et al.*, 1985; Cook and Struckmeyer, 1986; Khorasani 1987; Horsfield *et al.*, 1988) began to reconsider the long held view that coals could only source gas. Now, in the mid 90's, the majority of geochemists take a more open view on the role of coal and prefer to consider each case on their own merits with no preconceived notions on whether or not they can generate oil. Basins with terrestrially-derived hydrocarbons are known to occur throughout the world. A review of such basins has been given by Murchison (1987). More recently, Scott and Fleet (1994) edited a special publication on coal and coal-bearing strata as oil-prone source rocks consisting of a number of interesting articles, such as by Powell and Boreham (1994) and Scott and Fleet (1994) who highlighted the limit of our knowledge and a number of current problems related to oil generation and expulsion from coals.

Workers studying coals in the SE Asia region and Australia have revealed important findings regarding the association of coal measures and oil pools. Khorasani (1987) demonstrated that within the Surat Basin, Australia, the type of flora and certain favourable biochemical transformations (i.e. in dysaerobic conditions), in addition to other factors such as the environment of deposition and climate, could modify organic matter during burial and early diagenesis and so give rise to oil-prone coals of an autochthonous origin. Other workers (e.g. Thompson *et al.*, 1985) considered the accumulation of hydrogen-rich drifted coals and coaly sediments to have occurred by a separation process of water-soluble humic-acid gel from water-insoluble fragments of liptinitic components and humified wood in tidal dominated, coastal-plain environments. These kind of selective processes of degradation and reworking increases the content of hydrogen-rich kerogen and enhances the oil generating potential of coals and coaly shales.

The question of whether a coal can generate oil is not straightforward: a definitive "yes" or a definitive "no" should not be offered depending upon

which school of thought to which one belongs. Many issues have to be considered, such as which liptinitic maceral possesses the greatest potential; does the maceral vitrinite have a role to play: what is the relationship between organic microlithotypes and inorganic constituents, hence depositional environment, and how do they influence oil generation; at what maturation level is generation of oil from different macerals likely to occur; and what process/conditions are required for the effective expulsion of oil from coals. Many of these issues still do not have satisfactory answers but it is hoped that the Sarawak coals, due to their variety in type, setting and maturity, will provide some answers.

The coals currently under investigation are from the Bintulu area and the Merit-Pila coalfield, Sarawak (Fig. 1). These coals are Miocene in age (Wolfenden, 1960). In this preliminary study, the coals were analyzed in order to identify their common liptinitic constituents; to relate the liptinitic maceral types to oil generating potential; and to recognise features indicative of oil generation from coals. Coals from the Bintulu area were collected along the Bintulu-Miri road and the Bintulu Tatau road. The coal seams are generally thin, ranging from 20 to 30 cm in thickness although a single coal seam of over a meter in thickness was seen at Sungai Setiam. The Merit-Pila coals were

collected within the Stapang area. They consist of a number of core samples as well as outcrop samples from the Belawei-Mujun upper coal zone. The coal occurs in thicker seams compared to the Bintulu coals with some seams being more than 2 meter in thickness.

EXPERIMENTAL

Crushed samples of approximately 2 mm in size were mounted in slow-setting polyester ("Serifix") resin mixed with resin-hardener in mounting cups and allowed to set. Upon hardening, the blocks were ground on a water-lubricated diamond lap until the surface was flat and free of irregularities. The samples were then polished using successively finer grades of silicone carbide followed by alumina. Water is used as a lubricant for all these polishing stages.

Microscopical examination was carried out exclusively under oil immersion in plane-polarised reflected light using a Leica DMRXP microscope photometry system equipped with fluorescence illuminators consisting of BP 340-380 excitation filters, RPK 400 dichromatic mirror and LP 425 suppression filter. Maceral compositions were determined under both normal reflected "white" light and uv (ultraviolet) light. A total of 1,000

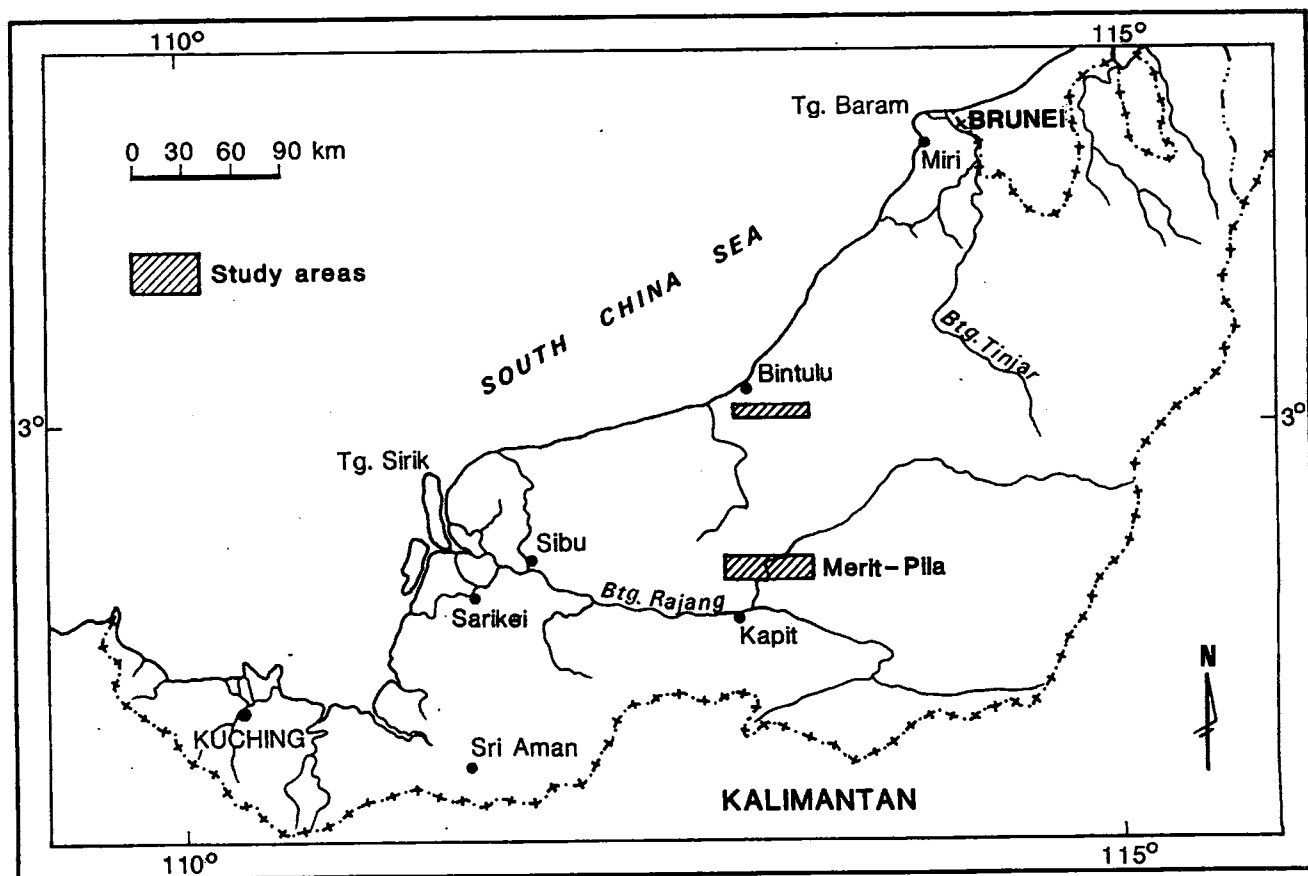


Figure 1. Location of the study areas.

point counts were made. Random vitrinite reflectance measurements ($\%R_o$) were carried out in reflected "white" light using a MPV SP photometer head. The GEOR Windows-based software package was used to acquire the vitrinite reflectance data.

RESULTS AND DISCUSSION

A summary of the vitrinite reflectance measurements ($\%R_o$) and maceral compositions for all of the samples analysed is presented in Table 1. A range of values is given for each parameter covering a total of fifty coals and a number of carbargilite samples that were analysed during the course of this study. The coals from the Bintulu area range in rank from sub-bituminous C to high volatile bituminous B. Coals of the Stapang area are relatively lower in rank, that is lignite to sub-bituminous C. Common liptinitic macerals in the Bintulu coals are shown in Figure 2 and that of the Stapang area is shown in Figure 3.

Petrographic features indicative of oil generation from coals

A number of petrographic features that are considered to indicate oil generation from coals have been recognized. Such features shown in Figure 4, include:

- i) the occurrence of exsudatinitic veins;
- ii) the occurrence of oil droplets/globules and oil haze;
- iii) changes of fluorescence intensity;
- iv) development of micrinite.

These features have also been recognized in coals from Australia and Kalimantan (Cook and Struckmeyer, 1986).

The most significant of these features is the occurrence of exsudatinitic (Fig. 4A), a secondary maceral commonly considered to represent the very beginning of oil generation in coal (e.g. Teichmüller, 1974). Drops of greenish-yellow fluorescing liquid (described as oil cuts by Cook and Struckmeyer, 1986), were seen to expel out from this maceral and some other liptinitic macerals (Fig. 4B). This oil-like material spreads out, leaving behind a thin film, recognizable by the presence of typical Newton's rings or blue staining (Fig. 4C). Besides these oil droplets, oil haze is the other common microscopic feature that was observed (Fig. 4A). It is formed as a result of volatilization of hydrocarbons, in this case the exsudatinitic, when in contact with the mounting resin.

An increase in fluorescence intensity is observed with hydrocarbon generation under uv light excitation (Fig. 4B). Once the hydrocarbon components have been expelled at higher thermal

maturation level, the fluorescence intensity of the precursor material and the exsudatinitic maceral decreases. The most distinct changes in the fluorescence with maturity is displayed by the exsudatinitic maceral. Exsudatinitic is typically a dark maceral as seen under reflected "white" light and fluoresces intense yellow-orange under uv light or "blue" light excitation (Figs. 2E and 4A). This distinct feature is observed in samples possessing vitrinite reflectance of about 0.45% in the Bintulu coals and 0.4% in the Merit-Pila coals. In coals of a relatively higher maturity of about 0.55% R_o , the exsudatinitic appear as a light-gray/brown staining in reflected "white" light (Fig. 2F), and in samples of approximately 0.7% R_o , the exsudatinitic appears whitish staining. Under uv light excitation, these relatively high maturity samples show a weak pale-orange fluorescence to virtually no-fluorescence. The decrease in the fluorescence intensity corresponds to the release of the liquid hydrocarbon by the exsudatinitic. Once all of the hydrocarbon components have been expelled, a micrinite-like substance may be formed (Fig. 4D). This is believed to be a disproportionation product as initially proposed by Teichmüller (1974) whereby the liquid bitumen is formed as one product and a residual solid substance, that is micrinite, as the other product.

The close association between the exsudatinitic maceral and vitrinite is shown in Figure 2E and clearly demonstrates that exsudatinitic is not merely a "crack-filling" maceral (i.e. filling in existing cracks), but rather develops the cracks and progressively forms a network as it extrudes out of the precursor macerals into the surrounding medium. Being soft and mobile at the time of exsuding, exsudatinitic also tends to migrate into pores and fissures as well as into cracks. The formation of exsudatinitic-crack networks is believed to represent an effective way of hydrocarbon expulsion from coal source rocks (Wan Hasiah, 1997).

Order of Generation

The oil-generative potential of a source rock is directly related to its content of hydrogen-rich organic matter. In coals, the hydrogen-rich components are mainly the liptinitic constituents such as alginite, bituminite, cutinite, resinite, sporinite and suberinite; whereby (with the exception of alginite), these are the common liptinitic constituents of the Tertiary coals currently investigated (see Table 1). Within the samples analysed, suberinite seems to possess the greatest oil-generating potential for coals of the Stapang area, whilst for coals/carbargilites of the Bintulu area, it is bituminite (Wan Hasiah, in press). This

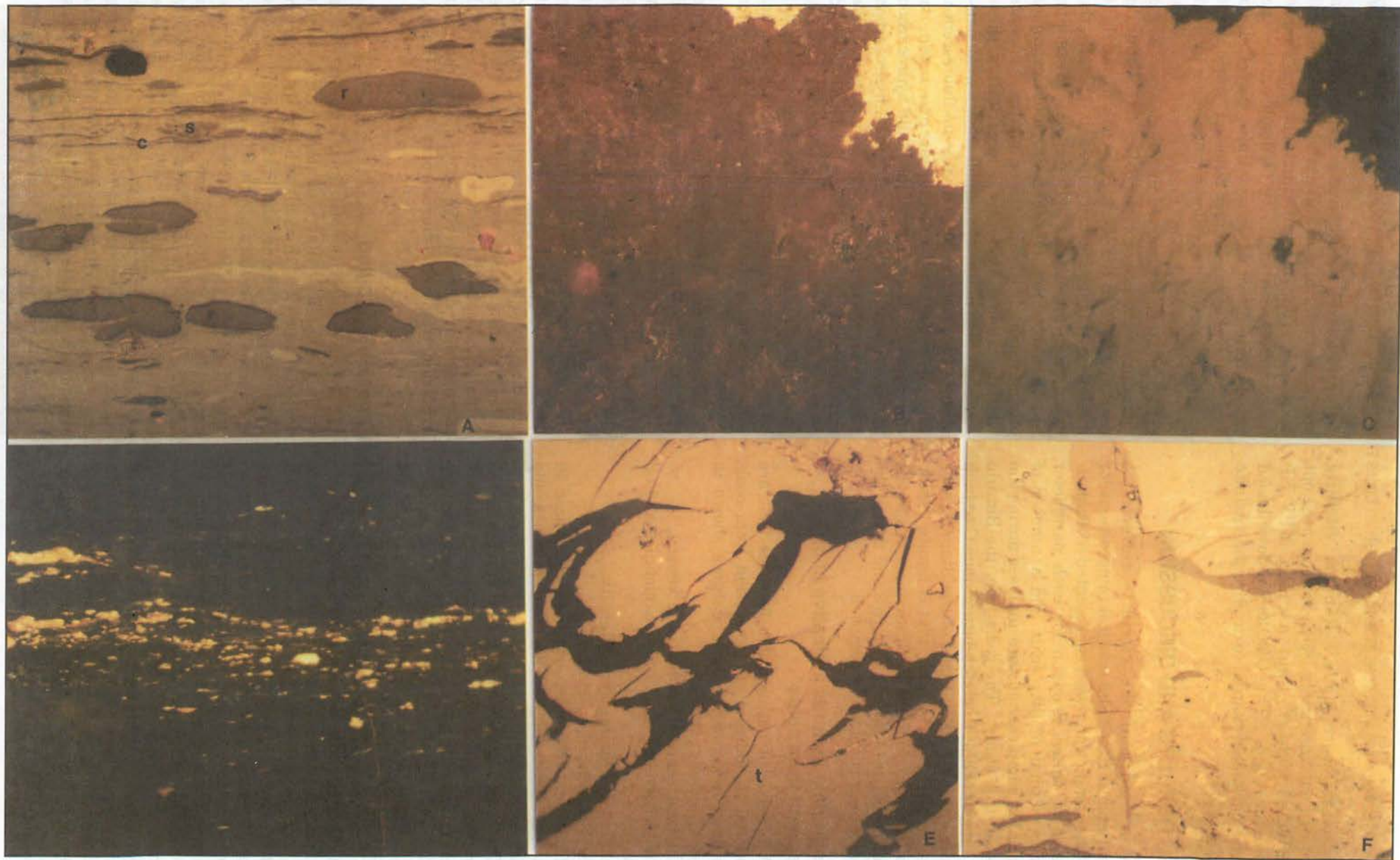


Figure 2(A-F). Common liptinitic macerals in coals of the Bintulu area (see opposite page for detailed descriptions).

Figure 2A. Dark gray resinite bodies (r) representing original cell excretions, associated with low occurrences of cutinite (c) and sporinite (s) in structureless vitrinite groundmass; reflected light, mag. 125x.

Figure 2B. Bituminite displaying characteristic properties of low reflectance, coloured internal reflections and lack of any characteristic shape or structure. Note the soft nature of this amorphous maceral demonstrated by the scratch marks formed during polishing; reflected light, mag. 125x.

Figure 2C. The amorphous nature of bituminite is enhanced under uv light irradiation under which it displays a pale yellow fluorescence (same view as Figure 2B).

Figure 2D. Fine grained lipid resinite and liptodetrinite fluorescing bright yellow to yellow-orange under uv light excitation (mag. 125x). Lipid resinite originates from fats and waxes often coating the surfaces of leaves and fruits whilst liptodetrinite are finely detrital liptinitic materials which may constitute fragments of other liptinitic macerals such as alginite, cutinite, sporinite etc.

Figure 2E. Dark brown veins of exsudatinitite entraining telocollinite (t) and forming a network as it extrudes into the surrounding medium (mag. 125x). Exsudatinitite is a secondary resinite, considered as a "solid bitumen" which presumably was soft and mobile while extruding. It represents the very beginning of bituminization and its genesis is related to the generation of hydrocarbons in coals; reflected light, mag. 125x.

Figure 2F. In coals of relatively higher maturity of about 0.6% vitrinite reflectance, exsudatinitite appears as light brown staining and shows a very vague fluorescence under uv light excitation. The decrease in the fluorescence intensity is believed to correspond to the release of the liquid hydrocarbons by the exsudatinitite maceral; reflected light, mag. 125x.

assumption is made based on their ability to generate significant quantities of the secondary maceral, exsudatinitite which is commonly considered to represent the very beginning of oil generation in coal (Teichmüller, 1974). The formation of petroleum hydrocarbons are generally considered to start generating in the sub-bituminous A coal stage, which is equivalent to about 0.5%R_o, while the actual expulsion begins later, in the high volatile bituminous coal stage, at about 0.7%R_o (Hood *et al.*, 1975; Tissot and Welte, 1984). However, recently, there have been a number of workers that suggest an early phase of hydrocarbon generation from coals that would normally be regarded as immature (e.g. Khorasani, 1987; Khorasani and Murchison, 1988; Zhao *et al.*, 1990; Khorasani and Michelsen, 1991; Wan Hasiyah *et al.*, 1995).

Within the samples studied, development of exsudatinitite is observed to be most extensively associated with the bituminite maceral, although minor occurrences of exsudatinitite associated with cutinite and sporinite are also observed in the Bintulu coals. For coals of the Stapan area the development of exsudatinitite is widely associated with the maceral suberinite and to a lesser extent with the maceral terpene resinite (i.e. dammar resin). No development of exsudatinitite is observed to be associated with sporinite, lipid resinite or other liptinitic macerals for coals possessing vitrinite reflectance < 0.6%. Based on the association and the abundance of these different types of liptinitic macerals at different maturation levels, it is possible to suggest that suberinite and terpene resinite are among the first liptinitic constituents of coals to generate liquid hydrocarbon, followed by bituminite and then cutinite. Subsequent generation of the oil-like material from the maceral sporinite and other resinous component (mainly lipid resinite) may be expected.

A distinct order of generation of hydrocarbons from liptinites has been recognised in an earlier work by Khorasani and Murchison (1988) based on the emission spectra of selected liptinitic macerals. Suberinite was observed to clearly generate earlier than the threshold of the defined "oil-window" of vitrinite reflectance 0.5%. Most of the other liptinitic macerals were noted by these workers to begin to generate in the early stages of the "oil-window", but alginite and lipid resinite generate relatively later than terpene resinite, cutinite and sporinite. This is very much in agreement with the observation made in this current study (with the exception of bituminite which was not analysed by Khorasani and Murchison, 1988), whereby, the order of liquid hydrocarbon generated by the liptinitic constituent currently studied are as follows:

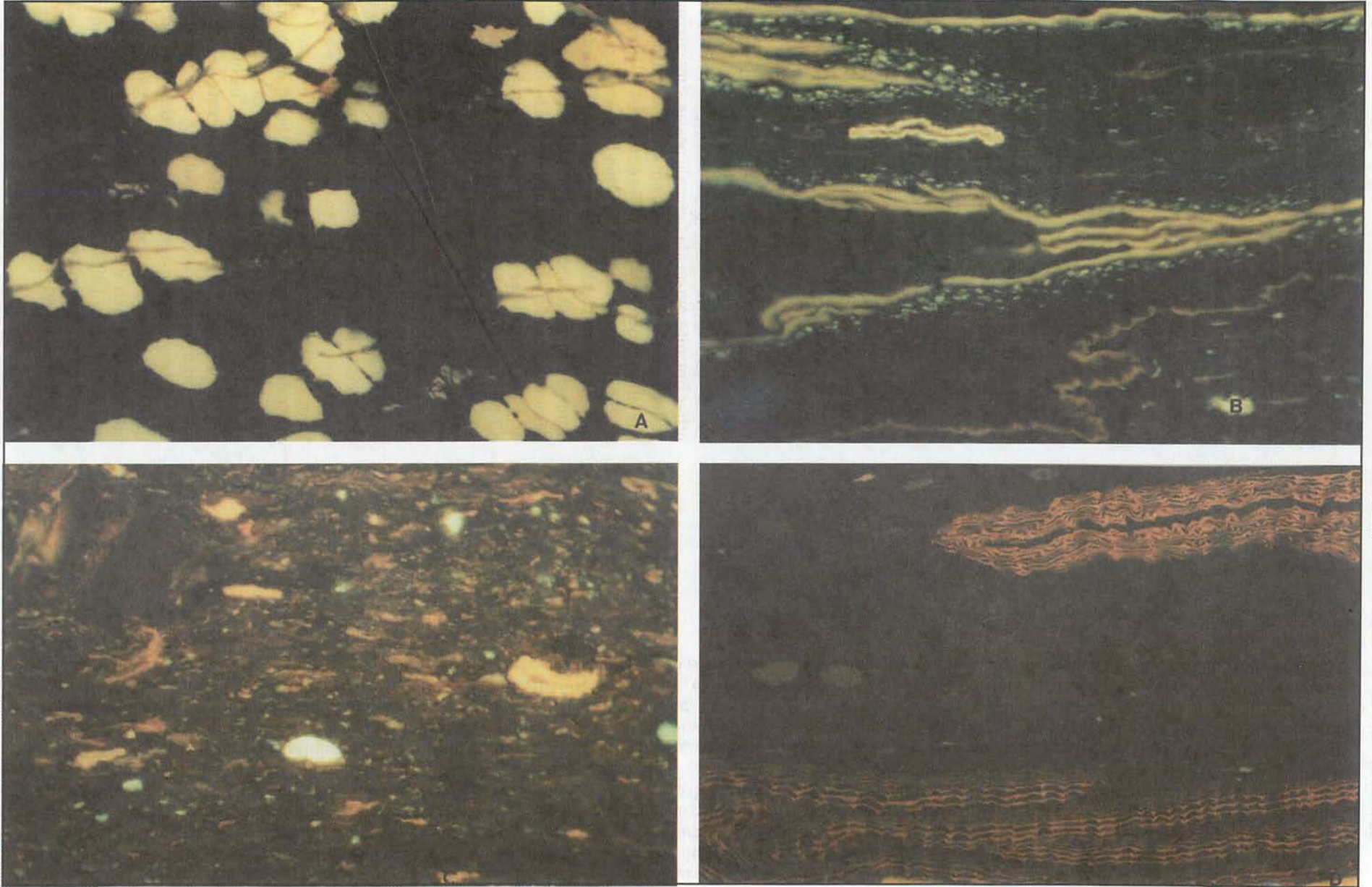


Figure 3(A-D). Common liptinitic macerals in coals of the Stapang ara (see opposite page for detailed descriptions).

Figure 3A. Terpene resinite of a dammar type resin considered to belong to a group of natural resins originating from angiosperm trees; uv light excitation, mag. 125x.
Figure 3B. Strands of yellow fluorescing thin-walled cutinite originating from cuticular layers of plant parts associated with whitish-green fluorescing fluorinite of probable plant oil origin; uv light excitation, mag. 160x.
Figure 3C. Yellow fluorescing sporinite (originating from the outer cell wall of spores and pollens) of predominantly thin-walled tenuisporites, although minor occurrence of thick-walled crassisporites also present. Closely associated with the sporinite is fine-grained yellow and green fluorescing liptodetrinite; uv light excitation, mag. 125x.
Figure 3D. Yellow fluorescing suberinite showing well-preserved cell walls of suberin-impregnated cork tissues. Cork tissue occurs mainly in barks and also at the surface of roots, on stems and on fruits, acting as a protection against desiccation; uv light excitation, mag. 125x.

Table 1. Summary of vitrinite reflectance (%R_o) and maceral composition (% by volume).

Blntulu (B) Samples	%R _o (random)	Common Liptinitic Macerals (%)					Other Liptinite ¹	Total Liptinite	Total Vitrinite	Total Inertinite	Mineral Matter
		Liptodetrinite	Resinite	Sporinite	Bituminite	Exsudatinite					
B — Miri Road	0.45–0.48	3–4	1–2	2–3	8–14	10–15	1–2	29–36	40–45	7–8	16–19
B — Tatau Road	0.56–0.72	2–3	1–20	2–7	trace–4	trace–4	1–2	12–31	55–78	3–9	3–11
Merit-Pila (Stapang) Samples	%R _o (random)	Common Liptinitic Macerals (%)					Other Liptinite ²	Total Liptinite	Total Vitrinite	Total Inertinite	Mineral Matter
		Fluorinite/ Liptodetrinite	Terpene resinite	Sporinite	Cutinite	Suberinite					
Outcrop	0.39–0.41	3–5	2–3	2–3	4–5	9–15	2–3	27–29	64–65	4–6	2–3
Core*	0.36–0.40	2–9	1–8	3–22	5–18	2–12	2–3	31–48	27–52	3–9	8–24

Other Liptinite¹: mostly cutinite

Other Liptinite²: mostly exsudatinite and alginite

Core*: With the exception of one sample having a very high abundance of terpene resinite (30%) and vitrinite (60%) associated with low occurrences of exsudatinite, inertinite and mineral matter.

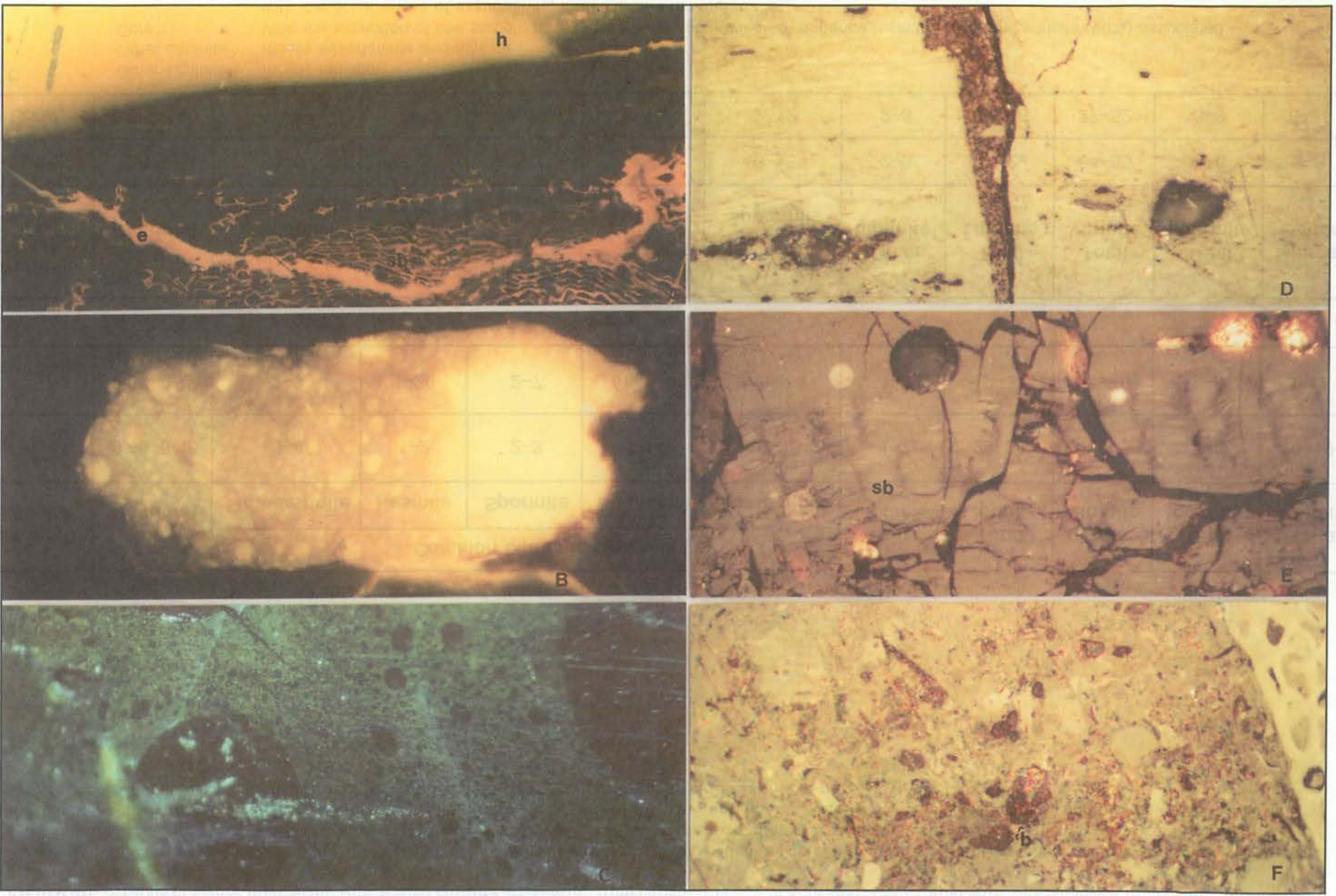


Figure 4(A-F). Petrographic features indicating oil generating from coals (see opposite page for detailed descriptions).

Figure 4A. Occurrence of exsudatinitic veins (e) originating from suberinite (sb) fluorescing yellow-orange and formation of oil haze (h). The latter is formed as a result of volatilization of hydrocarbons, in this case the exsudatinitic when in contact with mounting resin; uv light excitation, mag. 125x.

Figure 4B. Occurrence of oil droplets/globules in bituminite, and changes of fluorescence intensity is observed with hydrocarbon generation under uv light excitation, mag. 125x.

Figure 4C. Occurrence of oil staining whereby, under reflected "white" light, the oil globules appear as staining, normally blue in colour; mag. 125x.

Figure 4D. Development of micrinite once all of the hydrocarbon components have been expelled from exsudatinitic. Oil-like residue is still substantially present here but no fluorescence is observed when irradiated under uv light; reflected light, mag. 125x.

Figure 4E. Suberinite maceral (sb) showing a vague appearance in vitrinite groundmass (%R₀ of about 0.55%) as it has exhausted most of its hydrogen-rich components at an earlier maturation level; reflected light, mag. 125x.

Figure 4F. Parts of the bituminite macerals (b) have disappeared and formed the micrinite-like maceral surrounded by colourful oil stains in a carbargilite sample possessing vitrinite reflectance of about 0.55%; reflected light, mag. 160x.

	%R ₀ (vitrinite)
Suberinite/Terpene resinite	0.4
	↓
Bituminite	0.45
	↓
Cutinite	0.55
	↓
Sporinite/Alginate/Lipid resinite	> 0.6

The oil-like material is believed to be continuously generated by these liptinitic macerals within the range of maturity indicated above. Being among the earliest to generate, suberinite and bituminite (and to a lesser extent, terpene resinite) are also among the earliest to exhaust their liptinitic constituents. These macerals are observed to be "disappearing" in relatively higher rank coal (Figs. 4E and 4F) and thus possibly accounting for their low abundance in coals along the Bintulu-Tatau Road (see Table 1). The abundance of these liptinitic macerals and the maturation level attained by the host rock is considered among the most significant of factors that govern the oil-generating potential of coaly source rocks.

The observation made in this study agrees reasonably well with a number of suggested paradigms in relation to oil generation from coals by Cook and Struckmeyer (1986), in particular the efficient expulsion of oil-related compounds in low rank coals and the presence of labile material (i.e. exsudatinitic, in the present case) for coals to be effective source rocks for oil. No oil-like generation is, however, observed to be associated with the other two maceral groups (i.e. vitrinite and inertinite) within the coals currently investigated. Vitrinite, in particular telocollinite, merely provides an effective medium into which the hydrocarbon is expelled.

CONCLUSIONS

Based on this petrological study, it is clearly demonstrated that coals/carbargilites from the Bintulu and Stapang (Merit-Pila) areas are very rich in liptinitic materials. The common liptinitic constituents within the coals seem to vary between these two areas and this suggests variation in the original plant communities, type of deposition, depositional setting and possibly climatic condition also. The relative importance of these factors is presently being studied.

All of the coals and carbargilites analysed appear to possess a very high oil-generating potential based on the abundance of liptinitic constituents and their apparent capability to generate hydrocarbon as seen under the microscope. The most oil-prone maceral from the Bintulu area is regarded to be bituminite whilst that from the

Merit-Pila coalfield is suberinite. The exsudatinitite derived from these macerals represent an early phase of hydrocarbon generation which begins at about 0.4% vitrinite reflectance for suberinite and about 0.45% for bituminite. By about 0.7% vitrinite reflectance, the main bulk of the liquid hydrocarbon generated from exsudatinitite and their related precursors would have been expelled. However, the generation of liquid hydrocarbon is also to be expected from other liptinitite macerals such as cutinitite, sporinitite and alginite at different maturation levels than the bituminite and suberinite macerals.

It appears that one of the effective processes for expulsion of liquid hydrocarbon from coals and carbargilites is through the formation of exsudatinitite. Vitrinite, being commonly the most abundant macerals in coals, provides the medium into which the hydrocarbon, in the form of exsudatinitite, is being expelled. In clay-rich petroleum source rocks, where exsudatinitite seldom develops, the precursor material is believed to expel liquid hydrocarbon directly into the shaly medium.

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